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EFFECT OF HOT WATER EXTRACTION ON THE SOLUBILITY OF MILLED AND SOLID OAK WOOD (*QUERCUS ROBUR* L.)

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ABSTRACT

This study investigates the solubility of milled and solid oak wood (*Quercus robur* L.) samples using Method B (hot water extraction) as described in the ASTM D1110-21 standard. Serving as a continuation of previous research employing cold water extraction (Method A), the current work provides a comparative view of how elevated temperatures influence the release of water-soluble extractives, including tannins, gums, sugars, colouring matter and starches found in the wood. Although present in minor quantities, these extractives can significantly impact wood properties during hydrothermal processing, particularly in terms of discolouration and surface chemistry. The study focuses on oak wood (*Quercus robur* L.) due to its high technical and economic value in Croatia.

Keywords: penduculate oak (*Quercus robur* L.), hot water solubility, milled, solid, extraction, wood**.**

1. INTRODUCTION

Although they represent a relatively small proportion of the wood's chemical composition, extractives significantly influence its properties, including natural durability, colour changes during hydrothermal treatment, and aroma. The chemical structure of wood is complex and varies considerably among different species. On average, wood contains 46–56% cellulose, 23–35% hemicellulose, and 15–35% lignin. Extractives, non-structural components soluble in neutral solvents, typically constitute 4–10% of the dry mass of wood from temperate species but may account for up to 20% in tropical species. These compounds include a wide range of organic substances such as fats, waxes, alkaloids, proteins, simple and complex phenolics, sugars, pectins, mucilages, gums, resins, terpenes, starches, glycosides, saponins, and essential oils. Many serve as metabolic intermediates, energy reserves, or defence agents against microbial attack. Extractives contribute to various wood characteristics, including colour, odour, and resistance to decay (Pettersen, 1984).

Studies on the solubility of wood in organic solvents demonstrate that due to its heterogeneous chemical composition, different solvents are needed to extract specific molecular components. Most extractives can be removed using solvents such as water, benzene, toluene, or acetone. However, no single solvent is capable of extracting all components completely. The most effective extraction results of wood have been achieved using solvent mixtures, including acetone–water (9:1), ethanol–toluene (1:2), acetone–hexane, and ethanol–benzene (1:2) (Horvat, 2005). Water is particularly effective for extracting polar, low-molecular-weight compounds such as tannins, sugars, and certain phenolics. Hot water extraction (HWE) improves the solubility and diffusion of these substances from the wood matrix of some wood species. For instance, Dababi *et al*. (2020) successfully extracted tannins from Aleppo pine bark and sumac root using water medium, confirming its potential for isolating bioactive compounds for applications such as environmentally friendly adhesives.

The efficiency of water extraction depends on parameters such as temperature, extraction time, and wood species. Higher temperatures typically increase the yield of extractives, although they can also lead to degradation of heat-sensitive compounds. For example, Fang *et al*. (2013) reported that hot water extraction at 100 °C yielded 20 mg/g of extractives from spruce wood, while increasing the temperature to 140 °C raised the yield to 43 mg/g. Water extraction is considered an environmentally sustainable method, as it eliminates the need for organic solvents, making it particularly suitable for applications in food, pharmaceuticals, and bio-based materials. However, it is less effective for non-polar compounds such as fats, waxes, and resins, which require non-aqueous solvents, typically organic or certain inorganic solvents, for efficient extraction.

Water was selected as the solvent for this research because wood is frequently exposed to water during industrial processing, particularly during hydrothermal treatment. Additionally, water-soluble extractives play a crucial role in the winemaking industry. For example, barrels made from pedunculate oak are prized for their extractives, which dissolve into wine and contribute to its distinctive aroma, flavour, and colour.

2. AIM OF RESEARCH

The primary objective of this research was to investigate, through laboratory experimentation and subsequent analysis, the influence of drying and initial moisture content of pedunculate oak (*Quercus robur* L.), in both milled and solid form, on its solubility in hot water. Specifically, the study aimed to quantify the amount of extractive material obtained through hot water extraction (Method B), following the standard procedure outlined in ASTM D1110-21. The experiment was conducted at the Faculty of Forestry and Wood Technology, University of Zagreb. The study aims to determine how particle size and initial moisture content influence the efficiency of hot water extraction of wood extractives from milled and solid oak samples.

3. OBJECTS AND METHODS OF RESEARCH

3.1. Materials

Milled and solid samples of pedunculate oak (*Quercus robur* L.) were used to evaluate water solubility. The extraction was conducted using demineralised water (ASTM Type II), following the hot water extraction procedure (Method B) described in ASTM D1110-21 Standard Test Methods for Water Solubility of Wood. This standardised approach ensures consistency in determining the content of water-soluble constituents. Wood samples were sourced from oak lamellae with dimensions of 1000 × 130 × 7 mm, cut using an Einhell TC-SB 200/1 band saw. For the preparation of material intended for milling, each element was trimmed by 3 cm from both longitudinal edges and 7 cm from the end grain. The remaining central lamella (Figure 1) was further divided into samples weighing approximately 4 g and measuring 31 × 18 mm.

Figure 1. *Central lamella from which samples were made.*

Coarse milling was performed using a Retsch SM400 cutting mill with a 4 mm mesh screen, followed by fine milling with a Retsch SR300 rotor mill using 1 mm and 0.5 mm mesh screens (Figures 2 and 3).

Figure 2. *Retsch SM400 mill.*

Figure 3. *Retsch SR300 mill.*

The resulting material was sieved through laboratory-grade sieves with mesh openings of 425 µm and 250 µm. Only the fraction retained on the 250 µm sieve was used (Figure 4) for further analysis, while coarser and finer particles were excluded. For solid wood testing, elements were similarly trimmed by 3 cm on each lateral side and 7 cm from the end. Additionally, the central lamella was longitudinally reduced by 3 cm. Narrow slats were then cut from the remaining wood and used to prepare intact test specimens (Figure 5) as well as gravimetric samples.

Figure 4. *Milled material used.*

Figure 5. *Example of solid sample used.*

Solid wood specimens were cut to approximate dimensions of 20 × 15 mm to achieve a target mass of approximately 2 g. All samples, milled and solid, were air-dried prior to extraction to standardise moisture content.

3.2 Methods

The initial moisture content of the samples was determined by the gravimetric method using a Sartorius CPA225D analytical scale with a precision of 0.01 mg and a Memmert UF110 Plus laboratory drying oven. For solid wood samples, moisture content was determined using previously prepared samples, which were further cut into smaller specimens. For milled wood, the moisture content was assessed using excess material not required for other experimental procedures. Approximately 1 g of each sample was weighed into pre-dried containers and placed in the oven at 103 ± 2 °C until a constant mass was achieved. After drying, samples were transferred to a desiccator to cool to room temperature, after which they were reweighed. The moisture content of the wood was calculated using the following equation (1):

$$\omega = \frac{(W_1 - W_0)}{W_0} \times 100 \quad (1)$$

ω –moisture content (%)
W_1 – mass of the sample with moisture content (g)
W_0 – mass of the sample in an absolutely dry state (g)

The extraction process involved four sets of samples, each consisting of 6 replicates:

1. Milled samples with a defined moisture content
2. Solid samples with a defined moisture content
3. Milled samples in an absolutely dry state
4. Solid samples in an absolutely dry state

For the determination of hot water solubility, approximately 2 grams of each sample were weighed into a round-bottom flask (Figure 6). Subsequently, 100 mL of distilled water was added to the flask. The flask was then placed in a laboratory water bath equipped with a Liebig condenser (Figure 7). It was positioned such that the contents of the flask remained submerged below the water level in the bath. Extraction was conducted for a duration of 3 hours. After completion, the contents of the flask were filtered using the vacuum filtration system (Figure 8).

Figure 6. *Extraction aparatus.*

Figure 7. *Solid wood samples during extraction.*

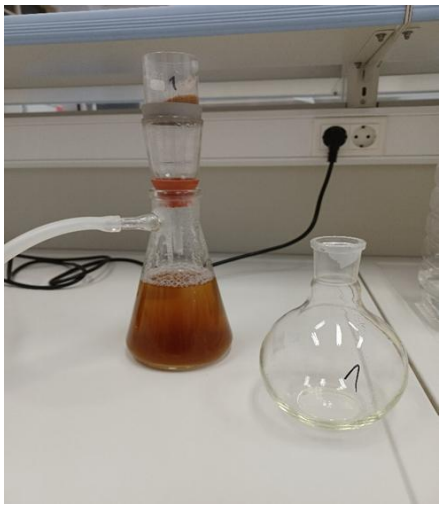

Figure 8. *Filtration process.*

The residue retained in the crucible was then dried at a temperature of 103 ± 2 °C using a Memmert UF 110 plus drying oven. The drying process lasted 4 hours. Upon drying, the samples were placed in a desiccator to condition at room temperature. Finally, the samples were weighed to determine the mass of the insoluble fraction.

4. RESULTS AND DISCUSSION

Results of descriptive statistics of solubility in hot water are shown in Table 1.

***Table 1**. Descriptive statistics of solubility in hot water [%].*

	Number of observations	Minimum	Maximum	Mean	Std. deviation
Milled samples (MC = 9.8 %)	6	6.49	10.28	7.5367	1.38228
Absolutely dry milled wood samples	6	6.78	8.20	7.5200	0.48683
Solid samples (MC = 11.34 %)	6	14.85	19.17	16.7850	1.57604
Absolutely dry solid samples	6	1.90	2.85	2.3467	0.31935

The descriptive statistical analysis gives information on how different physical states and moisture levels of wood samples influence their solubility in hot water. Four different sample conditions were examined: solid samples with 11.34% moisture content, completely dry solid samples, milled samples with 9.8% moisture content, and perfectly dry milled samples.

Among the tested groups, the solid samples with moisture content exhibited the highest mean solubility. This suggests that moisture plays a critical role in facilitating the release of water-soluble compounds, particularly in whole wood structures. The presence of moisture likely enhances the swelling of cell walls and increases the mobility of extractives, thus promoting higher solubility during the extraction process. However, this group also demonstrated the greatest variability among replicates, implying that the interaction between natural moisture and intact wood structure may introduce inconsistencies in solubility behaviour, possibly due to heterogeneous distribution of extractives or differences in wood density. On the opposite end of the spectrum, absolutely dry solid samples showed the lowest mean solubility and the most consistent results among all sample groups.

The lack of moisture in these samples reduces the swelling capacity of the wood matrix and limits the diffusion of extractable components into the surrounding solvent. The low variability within this group suggests a stable and predictable interaction between the dry solid structure and hot water, likely due to minimal structural changes during the extraction process. Milled samples, both with moisture content and in an absolutely dry state, exhibited intermediate levels of solubility. Interestingly, despite the reduction in particle size, which typically increases surface area and enhances extractability, the differences in solubility between moist and dry milled samples were marginal. This finding implies that while milling facilitates solvent access, moisture still plays a more dominant role in enhancing solubility. However, the milled samples with moisture content displayed greater variability compared to their dry counterparts, indicating that moisture content introduces additional factors affecting extractability, even in finely divided samples. Figures 9, 10, 11, and 12 show the distribution of results for each sample.

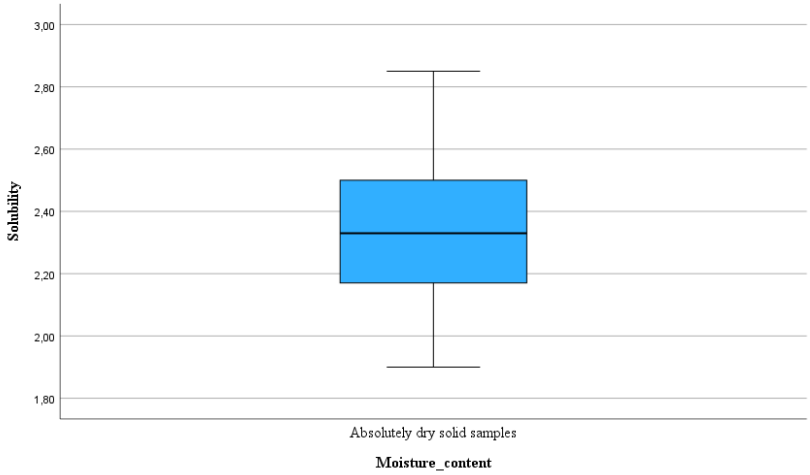

***Figure 9**. Solubility of absolutely dry solid samples [%].*

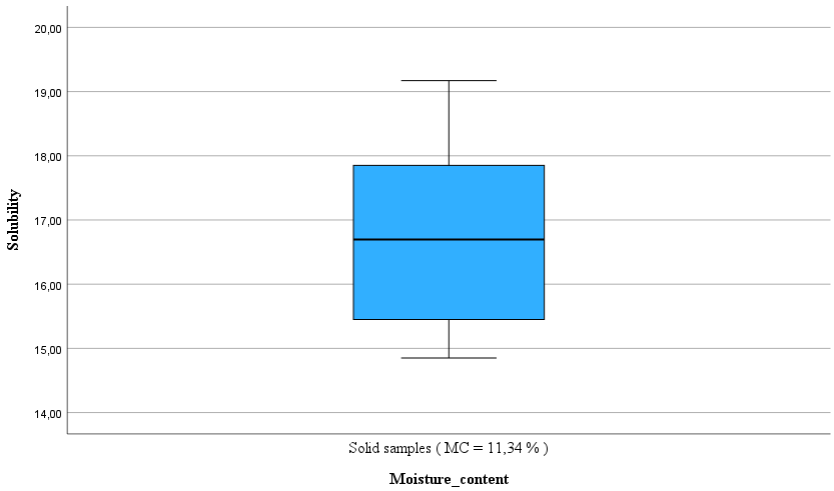

***Figure 10**. Solubility of solid samples with moisture content [%].*

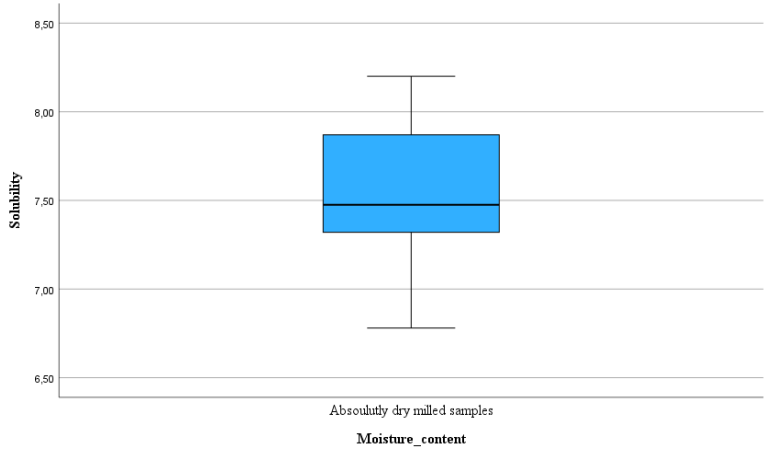

***Figure 11**. Solubility of absolutely dry milled samples [%].*

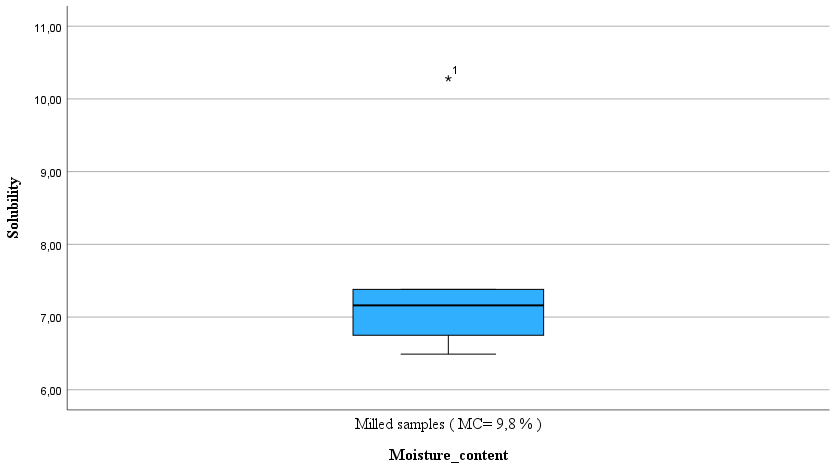

***Figure 12**. Solubility of milled samples with moisture content [%].*

Taken together, these results clearly illustrate that both the physical form of the sample and its moisture content significantly influence solubility outcomes. Moisture appears to have a greater overall impact than particle size reduction.

The Kruskal-Wallis test was performed to see if there were significant statistical differences between 4 groups of samples. The Kruskal-Wallis test showed there were statistical differences ($p <$

0.05), so post hoc pairwise comparisons of solubility in hot water were performed using the Mann–Whitney U Pairwise Comparisons (this test was chosen because of the non-normal distribution of the data and small sample size), with Bonferroni correction applied to account for multiple comparisons. (Table 2).

***Table 2**. Mann–Whitney U Pairwise Comparisons between samples.*

Sample Comparison	Test Statistic	Standard Error	Sig. (2-tailed)	Adjusted Sig. (Bonferroni)
Absolutely dry vs. absolutely dry milled samples	10.333	4.082	0.011	0.068
Absolutely dry solid vs. solid samples (MC = 11.34%)	18.000	4.082	<0.001	0.001
Milled samples (MC = 9.8%) vs. absolutely dry milled samples	-2.667	4.082	0.514	1.000
Milled samples (MC = 9.8%) vs. solid samples (MC = 11.34%)	-10.333	4.082	0.011	0.068

Among the six tested sample pairs, a statistically significant difference was observed only between the absolutely dry solid samples and the solid samples with a moisture content of 11.34% (adjusted $p = 0.001$). This significant increase in solubility suggests that higher moisture content prior to testing or the condition (milled or solid) of the sample during processing may enhance the leaching of water-soluble extractives or thermal degradation products when subjected to hot water. The result indicates that even moderate increases in pre-conditioning moisture can substantially influence the extractive behaviour of wood-based materials under thermal exposure. All other pairwise comparisons, including those between milled and solid samples or between absolutely dry samples with moisture content, did not yield statistically significant differences following Bonferroni adjustment ($p > 0.05$), although some unadjusted values suggested trends toward significance (*e.g.*, $p = 0.011$). These findings imply that factors such as mechanical processing (milling) or relatively small variations in moisture content (e.g., 9.8% vs. 11.34%) do not independently exert a strong enough influence on hot water solubility to be considered statistically meaningful. Overall, the results underscore the pronounced effect of the initial physical and moisture state of wood samples on hot water solubility, a parameter intimately linked to the presence of low-molecular-weight extractives, hemicellulose breakdown products, and residual chemicals from prior processing. In particular, the marked difference in solubility between fully dry and moisture-conditioned solid samples highlights the role of moisture in modulating both structural and chemical behaviours of lignocellulosic materials. These insights are especially relevant for optimising hydrothermal processing of wood, where moisture content and its interaction with thermal conditions affect the efficiency of steaming or drying treatments, ultimately influencing wood quality, dimensional stability, and colour change.

In comparison to the study conducted by Klarić et al. (2023), which measured the solubility of similar samples in cold water, the solubility of the samples in hot water was on average lower. This difference was more pronounced for milled samples than for solid samples, which exhibited similar average solubility values in cold water. Statistically significant differences in solubility were observed between milled and solid samples, both in the presence of moisture content and in absolutely dry conditions. Additionally, significant differences were found between solid wood samples with moisture content and those that were completely dry. However, when considering solubility in hot water specifically, a significant difference was observed only between absolutely dry solid samples and solid samples with moisture content.

5. CONCLUSION

- Moisture content of wood affects its solubility in hot water.
- Samples of solid wood with higher moisture content had higher average solubility than absolutely dry solid wood samples.
- In the case of milled wood samples, averages of solubility were similar between samples with moisture content and absolutely dry samples.

- In the case of samples that were absolutely dry, milled samples had higher average solubility than solid samples.
- Comparing it with cold water solubility, hot water solubility is on average lower than cold water solubility.

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